

University of Groningen

Optical properties of some half-metallic ferromagnets

Heide, P.A.M. van der; Baelde, W.; Groot, R.A. de; Vroomen, A.R. de; Engen, P.G. van; Buschow, K.H.J.

Published in:
Journal of Physics F%3A Metal Physics

DOI:
[10.1088/0305-4608/15/3/007](https://doi.org/10.1088/0305-4608/15/3/007)

IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.

Document Version
Publisher's PDF, also known as Version of record

Publication date:
1985

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Heide, P. A. M. V. D., Baelde, W., Groot, R. A. D., Vroomen, A. R. D., Engen, P. G. V., & Buschow, K. H. J. (1985). Optical properties of some half-metallic ferromagnets. *Journal of Physics F%3A Metal Physics*, 15(3). <https://doi.org/10.1088/0305-4608/15/3/007>

Copyright

Other than for strictly personal use, it is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license (like Creative Commons).

The publication may also be distributed here under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license. More information can be found on the University of Groningen website: <https://www.rug.nl/library/open-access/self-archiving-pure/taverne-amendment>.

Take-down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Downloaded from the University of Groningen/UMCG research database (Pure): <http://www.rug.nl/research/portal>. For technical reasons the number of authors shown on this cover page is limited to 10 maximum.

LETTER TO THE EDITOR

Optical properties of some half-metallic ferromagnets

P A M van der Heide[†], W Baelde[†], R A de Groot[†], A R de Vroomen[†],
P G van Engen[‡] and K H J Buschow[‡]

[†] Research Institute for Materials, Faculty of Sciences, University of Nijmegen,
Toernooiveld, 6525 ED Nijmegen, The Netherlands

[‡] Philips Research Laboratories, 5600 JA Eindhoven, The Netherlands

Received 12 November 1984, in final form 28 November 1984

Abstract. In this Letter we report ellipsometry measurements on polycrystalline samples of NiMnSb and PtMnSb at room temperature under ultra-high vacuum conditions. From these measurements the optical constants of the two materials were determined in the energy range 0.5–5.3 eV. The measured optical conductivities are in reasonable agreement with the optical conductivities calculated from the band structures by de Groot. The band gap for the minority-spin direction is clearly observed and can be brought into agreement with the band-structure calculations by consideration of spin–orbit splitting.

Considerable interest has focused on the Heusler (1903) $C1_b$ compounds NiMnSb and PtMnSb. The latter compound shows the highest magneto-optical Kerr effect of all materials at room temperature (van Engen *et al* 1983). A subsequent study of the electronic structure of these compounds by de Groot *et al* (1983) revealed a rather unusual situation. They found that both compounds are metallic for one spin direction while at the same time they show semiconducting properties for the opposite spin direction. This phenomenon was called half-metallic ferromagnetism. It is also related to the high magneto-optical Kerr effect in PtMnSb (de Groot *et al* 1984). Ellipsometry had already been performed on NiMnSb and PtMnSb by van Engen (1983). The main purpose of these measurements was to allow the calculation of the off-diagonal elements of the dielectric tensor complementing magneto-optical experiments. These measurements, however, were not performed under vacuum conditions and no effort was made to relate them to band-structure calculations. The main source of error in the values of the non-diagonal matrix elements δ of the dielectric tensor ϵ is inaccuracy in the values for the dielectric constants ϵ_1 and ϵ_2 . Therefore an independent determination of ϵ_1 and ϵ_2 makes a comparison with the values for δ found by van Engen (1983) possible.

The equipment used for measuring the optical properties consists of a photometric ellipsometer which, under ultra-high vacuum conditions, gives reliable results from 0.5–5.6 eV. This ellipsometer has been described elsewhere (van der Heide *et al* 1984a). Linearly polarised light is incident on the sample at an angle of 70°. The resulting intensities of the light reflected by the sample are measured at discrete positions of a rotatable analyser.

From these data the ellipsometric parameters $\tan \psi$ and $\cos \Delta$ (Beattie 1955) can be calculated and they enable us to determine the dielectric constants ϵ_1 and ϵ_2 . To compare the measurements with band-structure calculations, derived quantities were also

calculated—the most important ones being the optical conductivity σ and the joint density of states (JDOS).

The relation between ϵ_2 and these two quantities is given by

$$\text{JDOS} = A \sum_{i,f} \int dk \delta(E_f(k) - E_i(k) - \hbar\omega) = \omega^2 \epsilon_2 \quad (1)$$

$$\sigma = \omega \epsilon_2 \quad (2)$$

where i and f denote initial and final states and $\hbar\omega$ is the photon energy used.

The reflectivity and the energy loss function (E_{loss}) were also calculated. The energy loss function is defined by

$$E_{\text{loss}} = \epsilon_2 / (\epsilon_1^2 + \epsilon_2^2). \quad (3)$$

The spectra were recorded with photon energy increments of 0.01 eV up to 2.00 eV, of 0.02 eV between 2.0 and 2.4 eV and of 0.03 eV above this photon energy.

In recent measurements on Ni_3Al (van der Heide *et al* 1984b) the reproducibility of values of ϵ_1 and ϵ_2 for different samples which were polished in an identical way was shown to be 1%.

The samples were prepared by arc melting, using starting materials of 99.99% purity. X-ray diffraction was used to check the single-phase condition of the samples. To prepare an appropriate piece for our experiments the samples were spark-cut. The resulting spark-machined surfaces were polished with successively finer powders, ending with 1 μm diamond powder.

For NiMnSb this resulted in a perfectly shiny surface of 8 mm diameter. The measured deviation between the centre and rim of the sample was 0.27 μm , giving an optically flat sample. Laue photographs of the NiMnSb sample showed it to consist of a few grains only.

After polishing, the PtMnSb surface (about 32 mm^2 in size) showed a few holes (of diameter 60 μm) due to vapour inclusions and the presence of 1–2% of a second phase, which had escaped detection by x-ray diffraction. Laue photographs indicated that the sample was truly polycrystalline. Immediately after polishing, the samples were rinsed with pure ethanol and introduced into a turbomolecular pumped vacuum system, in which the measurements were performed at a pressure of 2×10^{-9} Torr.

Because both samples have a cubic crystal structure (Heusler Cl_b) the dielectric constants could be calculated using the formulae for isotropic samples (Born and Wolf 1959). The values of ϵ_1 and ϵ_2 are plotted in figure 1. Some structure can be seen in these graphs. As structures due to interband transitions are most clearly seen in the optical conductivity, this quantity was calculated from the measured data.

To separate the free-electron contribution (Drude term), which is only large in the infrared region, from the contribution due to interband transitions we calculated the plasma frequency ω_p and the scattering time τ , which determine the Drude behaviour. This was done by fitting an ideal Drude term to the measured ϵ_2 data in the low-energy region and by constructing a plot of $\hbar\omega\epsilon_2(\omega)$ against $-\epsilon_1(\omega)$ (Argand diagram).

For NiMnSb this resulted in $\tau = (1.4 \pm 0.2) \times 10^{-15}$ s and $\hbar\omega_p = (4.9 \pm 0.2)$ eV. For PtMnSb the values were $\tau = (8 \pm 1) \times 10^{-16}$ s and $\hbar\omega_p = (6.1 \pm 0.4)$ eV.

The optical conductivity calculated from a Drude curve with the above parameters was subtracted from the measured optical conductivity. These conductivities and the curves describing the difference between the fit and the measurements are shown in figures 2(a) and (b) for NiMnSb and PtMnSb respectively.

Qualitatively, the spectra of NiMnSb and PtMnSb look rather similar. The onset of

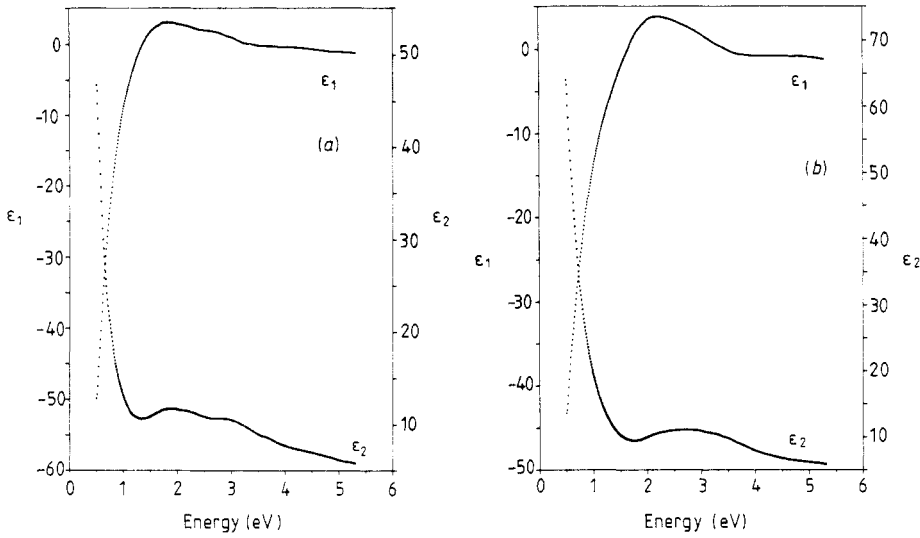


Figure 1. The real (ϵ_1) and imaginary (ϵ_2) parts of the dielectric constants from the measured data for (a) NiMnSb and (b) PtMnSb.

interband transitions occurs at 0.7 eV for NiMnSb in the optical conductivity region, after correcting for the Drude contribution. There is a shoulder at 2.00 eV and a broad peak at 3.20 eV. In the higher-energy range there is a small broad peak at 4.60 eV. For PtMnSb the onset of interband transitions occurs at 0.90 eV. The optical conductivity of PtMnSb also shows peaks at 3.40 eV and 5.16 eV.

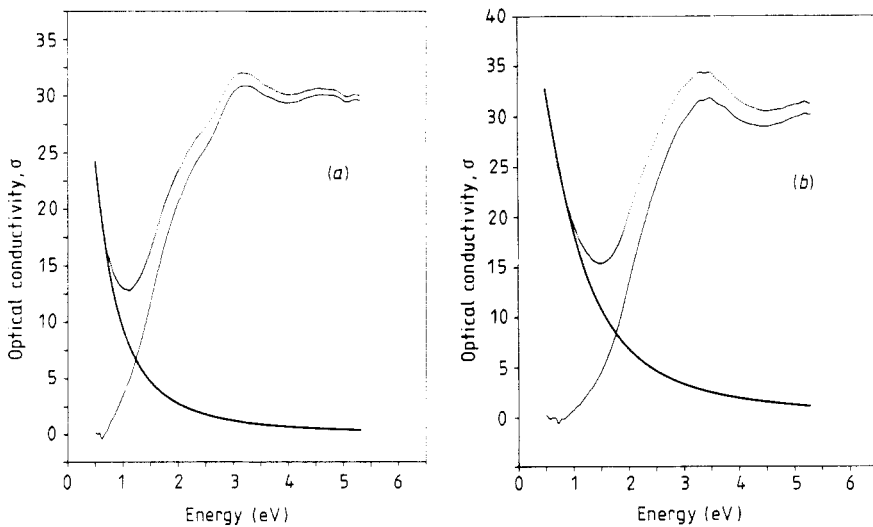


Figure 2. The optical conductivity σ of (a) NiMnSb and (b) PtMnSb from the experimental data (dotted curve). The thick full curve represents the optical conductivity as calculated for a Drude fit. The thin full curve is the optical conductivity as measured minus the optical conductivity calculated according to Drude.

The reflectivity as calculated from the measurements shows a rapid decline for both materials. For NiMnSb it reaches a minimum of 41% at 1.55 eV and for PtMnSb a minimum of 37% at 1.92 eV. At higher energies the reflectivity of NiMnSb varies between 38% and 42%. The reflectivity of PtMnSb varies between 35% and 42% at energies above 1.92 eV, with a local maximum at 3.4 eV.

The energy loss function shows a sharp peak for NiMnSb at 1.34 eV ($\epsilon_1 = 0$ at 1.36 eV), and for PtMnSb at 1.68 eV ($\epsilon_1 = 0$ at 1.66 eV). At these energies ϵ_2 is also small, and therefore these peaks are attributed to a plasmon being shifted in energy because of the interband transitions.

From our measured values of ϵ_1 and ϵ_2 for PtMnSb we also calculated the quantity $\text{Im}(\delta)$, using the values for Kerr rotation and ellipticity given by van Engen *et al* (1983). The energies of the maximum and minimum of the resulting curve are in good agreement with those given by van Engen (1983), but with respect to the magnitude of $\text{Im}(\delta)$ there are differences, particularly at energies below 2 eV. In this region our equipment can be considered more reliable than that used in the earlier work.

The optical conductivity was calculated from the band structures by de Groot (1983, 1984) using the same method as described before (van der Heide *et al* 1984a). The resulting theoretical optical conductivities for minority and majority spins are given in figure 3.

In both materials the majority spins only contribute to a relatively smooth, structureless background. In NiMnSb the threshold energy for the minority spins at 1.2 eV is seen clearly. This is followed by a peak at 1.95 eV which extends from 1.8–2.3 eV. In addition there is a steep peak at 3.03 eV which extends from 2.4–3.7 eV. In PtMnSb the contribution of the minority spins starts slowly at 1.0 eV and becomes stronger at 1.32 eV. There are also two broad peaks, one extending from 2.0–3.3 eV, the other extending from 4.3–5.0 eV.

The gross features of the optical conductivity are fairly well reproduced by the calculation. The peaks at higher energies (which all originate from transitions from states of primarily Sb character to Mn d states at various places in the Brillouin zone) are rather well reproduced, albeit with less structure than found with the calculation. This may stem from the assumption of constant matrix elements in our calculation of the JDOS. The peak between 4.3 and 5.0 eV for PtMnSb arises from transitions of Pt d to Mn d bands; the corresponding transitions for NiMnSb of Ni d to Mn d bands fall under the peak at 3.0 eV. The assumption of constant matrix elements and the fact that here mainly d to d transitions are involved would explain the differences between the experimental and the theoretical curves from 4.0–5.0 eV. The biggest discrepancy is in the position of the minority-band edge, which is found experimentally to be somewhat smaller in PtMnSb but considerably smaller in NiMnSb than the theoretical values. This discrepancy could well be due to the spin-orbit interaction, not included in the calculation of the JDOS. The spin-orbit interaction splits the top of the valence band Γ in the semiconducting spin direction into three equidistant singlets (de Groot *et al* 1984). The size of this spin-orbit interaction is about the same in both compounds, since the wavefunction character of the states at the top of the valence band is made of primarily Sb p states in both cases. The position of the Fermi energy in the two cases is different, however. In PtMnSb the Fermi level is situated only just above the top of the Sb-dominated valence band. Spin-orbit interaction makes the $m = +1$ state cross the Fermi energy and it thus ceases to be available as an initial state for an optical transition (this phenomenon is also related to the high magneto-optical Kerr effect in PtMnSb). In NiMnSb, however, the position of the Fermi energy is much higher in the gap and closer to the conduction band, and inclusion of the

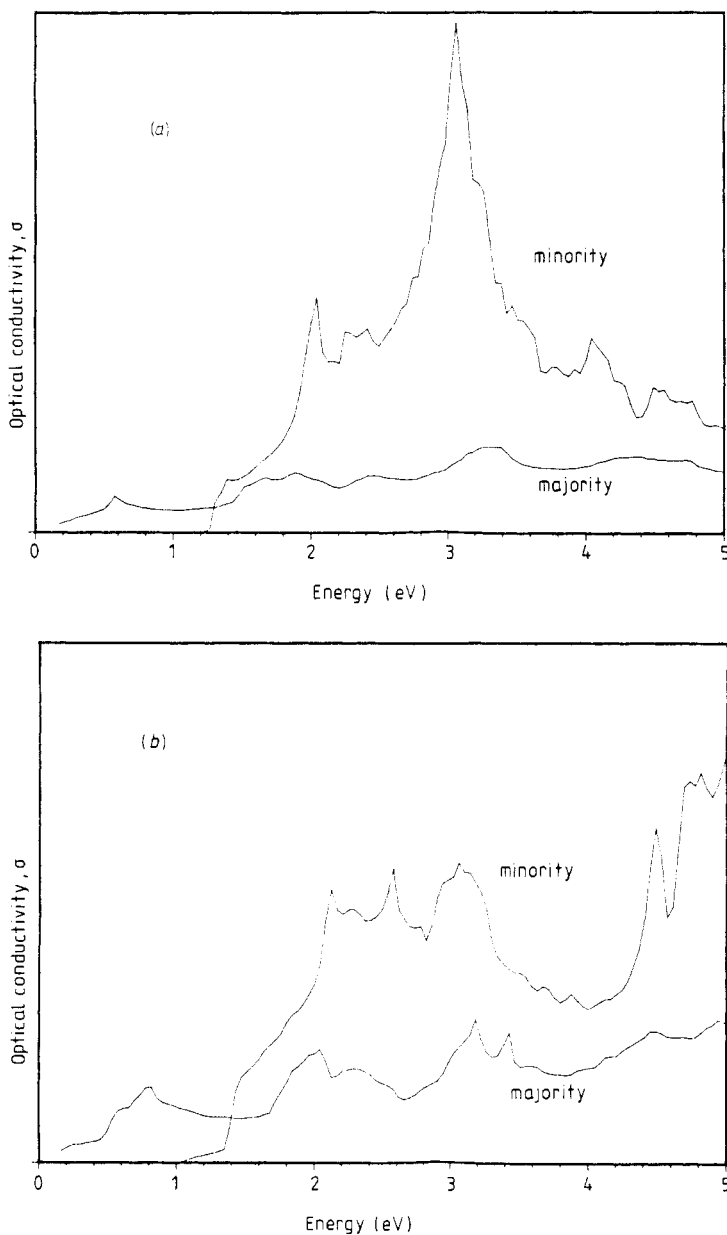


Figure 3. The optical conductivity σ of (a) NiMnSb and (b) PtMnSb calculated from the band structures for both minority and majority spins.

spin-orbit interaction will not then lead to an overlap of valence band and Fermi energy. The consequence is that the spin-orbit interaction would reduce the gap in NiMnSb by 0.25 eV while the effect in PtMnSb should be considerably less. This explains the different behaviours of PtMnSb and NiMnSb in the measured data but gives no explanation for the remaining difference of 0.2 eV for the onset of the interband transitions. The remaining difference of 0.2 eV is of the order of the experimental accuracy of the determination of

the gap. Also the local density approximation and relaxation effects could explain the difference.

The authors would like to acknowledge the assistance of L W M Schreurs and H M Weijers in the preparation of the samples for the ellipsometric experiment. Part of the work described was supported by the research programme of the 'Stichting voor Fundamenteel Onderzoek der Materie' with financial support from the 'Nederlandse Organisatie voor Zuiver Wetenschappelijk Onderzoek'.

References

- Beattie J R 1955 *Phil. Mag.* **46** 235
Born M and Wolf E 1959 *Principles of Optics* (Oxford: Pergamon)
van Engen P G 1983 *Thesis* Technological University Delft
van Engen P G, Buschow K H J, Jongebreur R and Erman M 1983 *Appl. Phys. Lett.* **42** 202
de Groot R A, Mueller F M, van Engen P G and Buschow K H J 1983 *Phys. Rev. Lett.* **50** 2024-7
—— 1984 *J. Appl. Phys.* **55** 2151
van der Heide P A M, Baelde W, de Groot R A, de Vroomen A R and Mattocks P G 1984a *J. Phys. F: Met. Phys.* **14** 1745-50
van der Heide P A M *et al* 1984b to be published
Heusler F 1903 *Verhandl DPG (VI)* **5** 219